

Scanning tunneling measurements of potential steps at grain boundaries in the presence of current flow

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We have used a new technique to simultaneously measure the surface topography and surface potential of current-carrying polycrystalline $\text{Au}_{60}\text{Pd}_{40}$ thin films using a scanning tunneling microscope. We find abrupt steps in the surface potential due to scattering from grain boundaries in these films.

Introduction

In the first volume of this journal over 30 years ago, Landauer [1] presented a picture of transport in metals which was in contrast to the conventional Boltzmann picture. In the new picture the resistance of the metal is ultimately the result of a series of small voltage drops

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occurring when the carriers collide with imperfections in the lattice potential. In particular, this paper [1] described deviations from the average gradient in potential which take the form of a dipole potential for the case of a point scatterer, or a step in potential for the case of a planar (e.g., grain boundary) scatterer. In the conventional Boltzmann picture, the details of the impurity potential are ignored completely or removed from the problem by averaging, and consequently the potential is taken to have a constant gradient along the sample. The picture of localized changes in the carrier potential has gained wide acceptance among theoreticians [2]. The theoretical description of quantum-mechanical effects in the conductivity of metals has relied on models which incorporate the local fluctuations in the chemical potential which result from impurity scattering [3, 4]. Theoretical descriptions of electromigration effects in metals also incorporate these effects [5]. To date, experimental confirmation of these assumed microscopic potential fluctuations has been indirect. We describe in this paper direct experimental observations of such fluctuations [6].

For these experiments, we used a method somewhat different from that of Murali and Pohl [7], who were the first to use a scanning tunneling microscope [8] as a potentiometer. Our method instead took advantage of the interrupted-feedback technique of Feenstra, Fein, and coworkers [9, 10]. At voltages low in comparison to the tunneling barrier potential (typically a few electron volts), the tunneling current-voltage characteristic between two metals is linear. An example of this is shown in Figure 1, which displays an experimental current-voltage characteristic between a Au-Pd film and a Pt-Rh tunneling tip. These data were taken at room temperature in vacuum. This current-voltage characteristic does not pass through the origin, because externally applied transverse current through the film has changed the potential directly below the tip relative to ground. In our method we infer the local potential of the sample by measuring the current I_0 at zero bias. Since the current-voltage characteristic is linear, and since we know the current both at the feedback point and at zero bias, we can infer V_s , the bias voltage which would give us no current between the tip and sample. This is the bias voltage at which the tip and sample are at the same potential. In our method (see the insert of Figure 1) the tunneling tip is connected to virtual ground through a current-sensitive preamplifier. The sample is biased with a square-wave voltage train through a bridge configuration which allows us to adjust the local potential between the tunneling tip and the sample. In the portion of the cycle when the sample is biased at a voltage V , the feedback loop to the z-piezoelectric is held closed, and the surface topography is measured in the standard way [8]. The feedback loop is then interrupted, the sample bias is brought to zero, and the tunneling current, from which we infer the surface potential V_s , is measured. The tip is then moved to a different point on the film, and the cycle is repeated to simultaneously measure the surface topography and potential. As can be seen in Figure 1, there is progressively more noise at larger tip-sample potential differences. Therefore, the potential signal is substantially less noisy when the tip-sample potential is close to zero [6]. This is the underlying advantage of this technique over that of Murali and Pohl [7].

The samples were mounted in a single-piezoelectric-tube, differential-spring-approach scanning tunneling microscope, which has been described in detail elsewhere [10]. All of the scanning tunneling microscope measurements reported here were made in vacuum at room temperature. The measurements were made on 60% Au, 40% Pd ($\text{Au}_{60}\text{Pd}_{40}$) films sputtered onto oxidized silicon wafers through metal shadow masks. Two silver-paint contacts defined the length of the film, which was 450 μm long, 220 μm wide, and 600 Å thick. The films had rather high resistances of about 350 $\Omega/\square$, and resistivities of about 2000 $\mu\Omega\text{-cm}$. In spite of the large values of resistivity, the films are good metals—the film

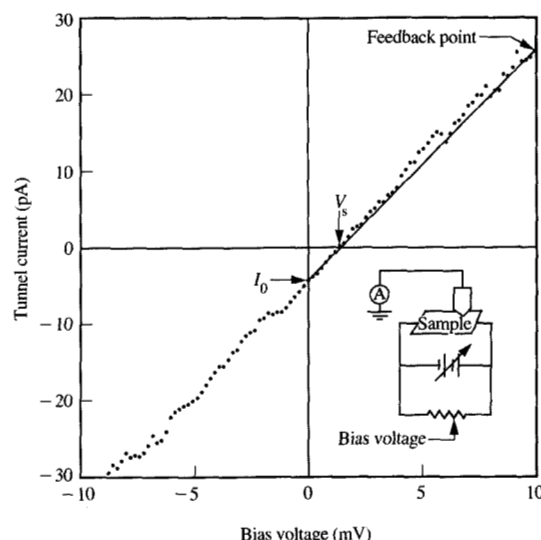


Figure 1

Current-voltage characteristic for tunneling into our $\text{Au}_{60}\text{Pd}_{40}$ films from a Pt-Rh tip at room temperature. The inset gives a diagram of our biasing technique.

resistance is far below the metal-insulator transition value, and the resistance is nearly independent of temperature (decreasing by a few percent by $T = 5$ K) and independent of the current through the film. Estimates of mean free paths from the resistivities of these films yield unrealistically short values (shorter than the lattice spacings). In contrast, estimates for the reflectivity of the grain boundaries from Landauer's formalism [1], given the measured film resistivities, yield values close to 1. The self-consistent picture can then be drawn in which the resistance of the films is dominated by grain boundaries which scatter electrons strongly. This picture is supported by our potentiometric measurements.

Figure 2 shows topographic (left column) and potentiometric (right column) perspective view images acquired simultaneously using the technique described above for one of our films. In Figures 2(a) and 2(b) a transverse electric field along the film of $E_t = 85$ V/cm was applied, with the scan and current directions oriented in such a way that the average voltage drop [indicated by the arrow on Figure 2(b)] was from right to left. Figures 2(c) and 2(d) were taken with the much smaller average field of 2 V/cm across the film from right to left. This small field of 2 V/cm was used to compensate for small residual voltages in the experimental apparatus, so that the tip-sample potential drop was as close to zero as possible. All of the images

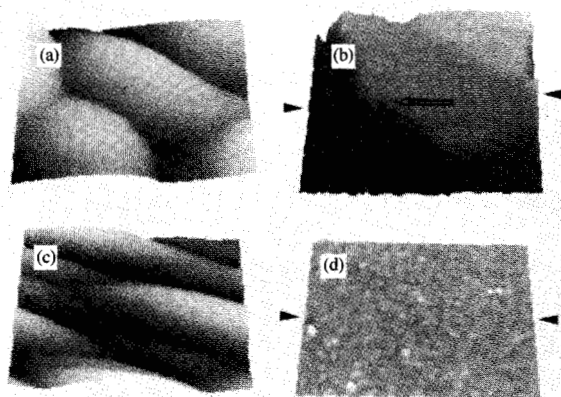


Figure 2

Topographic (left column) and potentiometric (right column) images of $\text{Au}_{60}\text{Pd}_{40}$ film under fields of 85 V/cm and 2 V/cm, respectively. Each pair of topographic and potentiometric images was taken simultaneously. Because of thermal and piezoelectric drifts, each pair of images was of a different region of the film. The topographic images are grey-scale images with 237 Å and 184 Å from white (high) to black (low) for (a) and (c), respectively. The potentiometric images [(b) and (d)] are on a grey scale of 650 μV from white (positive) to black (negative).

covered areas 240 Å high by 250 Å wide. The grey scales for the topographical images [2(a) and 2(c)] are 237 Å and 184 Å, respectively, from white (high) to black (low). The scales for the potentiometric images of the sample [2(b) and (d)] are 650 μV from positive (white) to negative (black) with respect to the tip. All of these images were recorded with a square-wave amplitude of $V = 1$ mV and currents of 100 pA in the half of the cycle in which the feedback loop was on. Similar results, with progressively poorer signal-to-noise ratios, were obtained for square-wave amplitudes up to 20 mV.

The histograms of Figure 3, which plot the frequency of occurrence vs. measured surface potential, show clearly that when current flows there are well-defined plateaus in V_s , while Figure 2 shows that each plateau corresponds to individual grains in the topographic images. When much smaller currents flow through the film [Figure 2(d)], there is essentially no variation in the measured surface potential, and the histograms have a very narrow distribution. The full width at half-maximum of the histogram corresponding to small currents through the films is about 23 μV . It is the very small noise level afforded by our biasing technique that allows these small potential steps to be measured. The fact that we do not observe potential steps when there is little current flowing through the sample leads us to conclude that

we are not seeing effects due, for example, to charging or variations in contact potential between the tip and sample.

In Figure 4 we show cross sections through the potential images of Figures 2(b) and 2(d) along the cuts marked by the arrowheads. Those cross sections are labeled in the figure by the average fields across the films ($E_f = 85$ V/cm and 2 V/cm, respectively). Also included in this figure are dashed curves showing the relative potential variations that would be expected from the average field across the sample. The zero in potential in each of the experimental curves was set by the measurement bridge. It can be seen clearly from these figures that the bulk of the voltage drops occur at spatially well-defined points (corresponding to the grain boundaries in the topographical images of Figure 2). The local electric fields are an order of magnitude smaller inside the grains than at the grain boundaries. The total potential drops across the images have the correct sign and approximately the correct amplitude expected from the average electric fields across the films.

The expected rounding of the potential steps from the space charge near the barriers [1] should decay on lengths of the order of the screening length. We estimate that the screening length for our film is $\lambda_s \sim 4$ Å, which is near the

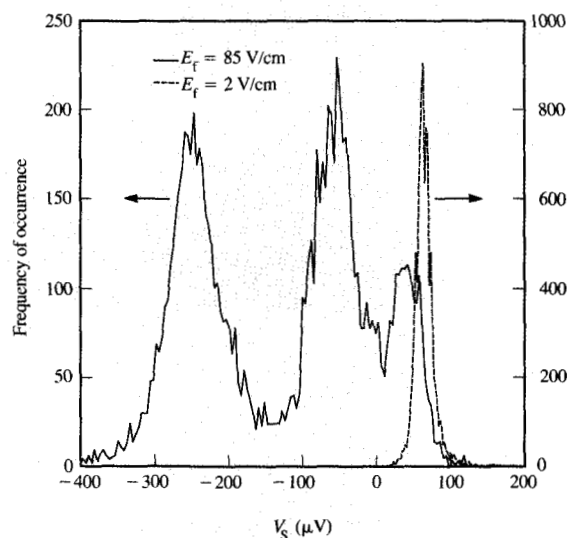


Figure 3

Histograms of the frequency of occurrence of each measured potential as displayed in Figures 2(b) and 2(d). When there is almost no current through the sample ($E_f = 2$ V/cm), the measured potentials have a sharp distribution. When a large transverse field is applied ($E_f = 85$ V/cm), there are distinct plateaus in potential corresponding to individual grains in the topographic image [Figure 2(a)].

lateral resolution of the microscope. There is some rounding of the steps in Figure 4 on length scales of about 5 Å, but given the noise level and the rough lateral resolution, we cannot conclude that these are the signatures of space charge near the grain boundaries. The traces in Figure 4 bear a strong resemblance to predictions for transport through a one-dimensional disordered medium [11] in the case where the scattering events are independent. (See, for instance, Figure 9 of [11].) The assumption that the scattering events are independent is appropriate for high temperatures where the phase memory length L_ϕ of the carriers is short. At lower temperatures, when L_ϕ grows to be larger than the average grain size, the scattering events will no longer be independent. Interference among the carrier wavefunctions will lead to correlation in their motion, and according to recent theory [4], the correlations in carrier motion have a length scale L_ϕ . We expect that this correlation will change the surface potential profiles in our films, and experiments to study this temperature dependence are currently underway.

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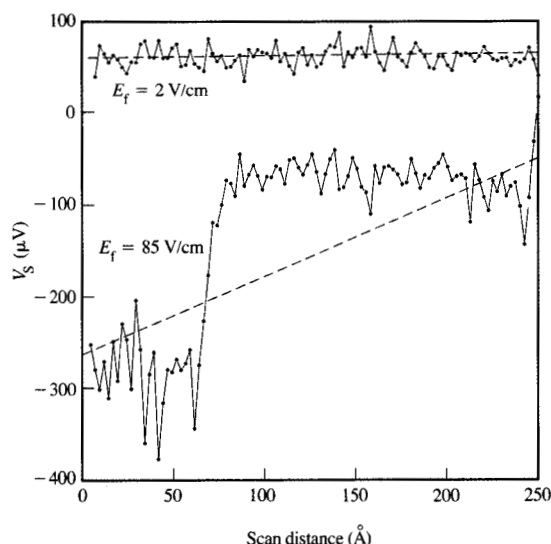


Figure 4

Cross-sectional cuts along the center lines (marked by the arrowheads) of the potentiometric images of Figures 2(b) and 2(d), corresponding to transverse film fields E_f of 85 V/cm and 2 V/cm, respectively. The dashed lines represent the expected potential drops from the average fields across the sample.

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